Response to the Reviewer #3

The manuscript uses 11-year measurements of BC at a single site downwind of China to estimate emission trends of BC in different regions of China. Meteorological variability was estimated based on the WRF-CMAQ model with fixed emissions. Although only one site is used, the 11-year-long data record has valuable information that can be used to constrain source emissions.

We appreciate the reviewer's positive comment on our manuscript. Detailed point-by-point responses are given below.

1) My main concern is on the robustness of the regional trends derived with this single site. The regional trends were estimated by sampling observations and WRF-CMAQ model outputs by the footprint of HYSPLIT trajectories (c.f. Figure 5). Neither WRFCMAQ model nor HYSPLIT footprint was evaluated with regards to their respective ability to simulate interannual variability of meteorology, particularly precipitation. What is the resolution of the GDAS1 meteorology to drive HYSPLIT? The manuscript does not specify the resolution; it might be as coarse as 1degree x 1 degree. Since the authors used two models, they should be cross-validated. For validation of the HYSPLIT footprint, the footprint for one region should be compared to WRF-CMAQ simulation with that region's emissions turned off to validate if the two methods provide consistent variability of BC at the receptor site.

The horizontal resolution of the GDAS1 meteorology to drive HYSPLIT backward trajectories was 1×1 degree. This information will be included in the revised manuscript. The resolution is similar to that used in the WRF/CMAQ model (ca. 80 km). As the NCEP Final Analysis data (ds083.2), on which the WRF model is based, originated from GDAS, high compatibility between the HYSPLIT backward trajectories and WRF/CMAQ was expected by nature.

A quantitative discussion on the influence from precipitation on our analysis will be included as follows, using the ΔBC/ΔCO ratio (Kanaya et al., 2016), for both observations and model simulations, to conclude that the influence is negligible, due to the adopted criteria (APT < 1 mm):

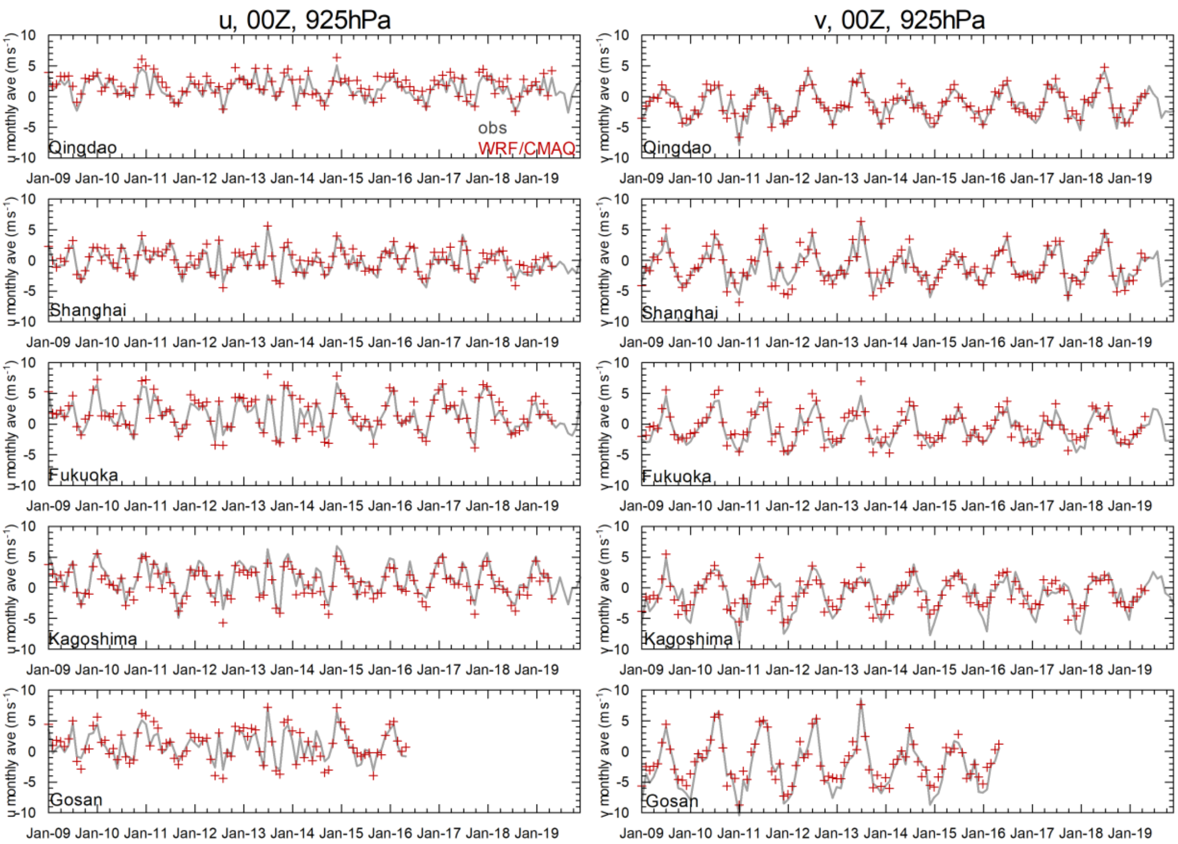
" The median ΔBC/ΔCO ratio for the observational data with APT < 1 mm (N = 26423) was only 1.9 % lower than that for data with APT = 0 mm (N = 18907), suggesting insignificant influence." (Page 4, Lines 33- Page 5, Line 2, in the track change document)

" Wet deposition was represented with the cloud_acm_ae5 module. Similarly to the observational data, the modeled ΔBC/ΔCO ratio decreased with APT; the modeled median ΔBC/ΔCO ratio for data with APT < 1 mm (N = 26737) was 3.7 % lower than that for data with APT = 0 mm (N = 19197). The removal in the model appeared stronger than the observational trend (1.9 %) but the error introduced due to the wet deposition representation was estimated to be small (−2 %) when using the adopted criteria (APT < 1 mm)." (Page 5, Lines 14-18)

Thus the model's reproducibility of the wind field and its interannual variability is of main concern. Figure S3 (as follows, to be included in the revised supplementary material) compares the observed and modeled (i.e., WRF) monthly-averaged zonal and meridional wind speeds during the whole study period at 925 hPa, over 5 meteorological observatories surrounding the Fukue site, i.e., over Qingdao, Shanghai, Fukuoka, Kagoshima, and Gosan. The absolute wind speeds and their month-to-month and interannual variations agreed quite well.

Nonetheless the origin region assignment will have uncertainty due to incompleteness of the used meteorological field. In the revised manuscript, uncertainty in the region assignment will be evaluated against trajectories based on the ERA5 meteorological field with a finer horizontal resolution (30 km) during 2013−2015, and the following statement will be included:

"While the assignment agreed for large fractions (83−93 %) on a country level (i.e., China Korea, and Japan), the successful fraction decreased to 57−67 % for the four individual Chinese regions, due to crosstalk with the adjacent regions." (Page 4, Lines 25-29)



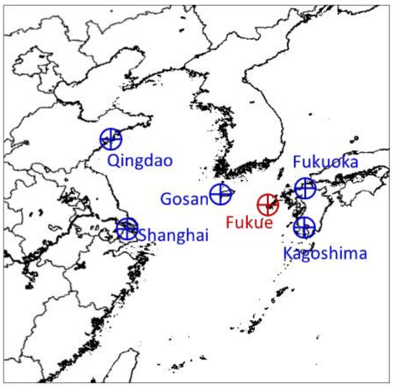


Figure S3. Observed monthly-averaged zonal (u) and meridional (v) wind speeds at 925 hPa at 00Z over five locations near Fukue (grey lines) were compared with those at corresponding girds of WRF/CMAQ model simulations.

2) For the validation of WRF-CMAQ, although the manuscript did compare WRF-CMAQ model against the EMeRGe-Asia campaign, that comparison was only for one spring season (2018) while the main use of the model was to estimate interannual variations of BC. It is the WRF-CMAQ model's ability to simulate interannual variability of meteorology that should be evaluated. For example, the manuscript should present WRFCMAQ simulated meteorological parameters at Fukue Island with those observed, and such comparison should be done for interannual scale. If data are available, the WRFCMAQ model's meteorology over mainland China should be evaluated too.

We will include the time-series plots comparing BC and CO observations with the WRF-CMAQ results for the whole study period (2009–2019) as Figure S1 in the revised supplementary material. There we demonstrate that individual peaks were well captured by the model not only during spring 2018 but also during the whole period. The reproducibility of the interannual variability of meteorology (wind field) was verified (shown above) and will be demonstrated as Figure S3 of the revised supplementary material.

We thank the reviewer again for the productive comments.

References

Kanaya, Y., Pan, X., Miyakawa, T., Komazaki, Y., Taketani, F., Uno, I., and Kondo, Y.: Long-term observations of black carbon mass concentrations at Fukue Island, western Japan, during 2009–2015: constraining wet removal rates and emission strengths from East Asia, Atmos. Chem. Phys., 16, 10689–10705, https://doi.org/10.5194/acp-16-10689-2016, 2016.